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## **EUROPEAN PATENT SPECIFICATION**

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(73) Proprietor: **COURTAULDS FILMS (HOLDINGS) LIMITED** Bridgwater Somerset TA6 4PA (GB)

(72) Inventor: Foulkes, Jonathan Paul Thornbury, Bristol BS12 2DS (GB)

(74) Representative: Claisse, John Anthony, Dr. 97 Portway Wells Somerset BA5 2BR (GB)

(56) References cited: EP-A- 0 143 567

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This invention concerns in-mold labels and articles having such labels applied thereto.

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In-mold labelling is a technique in which a pre- 5 formed polymeric label is incorporated into a polymeric article by molding the article in a mold containing the label. This technique in general provides labelled articles having a good appearance with the advantage that the labels effectively become an integral part of the articles and are thereby difficult if not impossible to remove. However, the molding process in general involves the use of high temperatures to effect melting or softening of the polymer used to form the articles, and since the pre-formed labels are usually made from polymeric films which have been stretched in their production, the temperatures to which the labels are subjected during the molding process usually lead to their shrinkage.

Although the dimensional changes resulting from label shrinkage can often be tolerated, and indeed anticipated when the labels are produced, shrinkage can lead to the films acquiring an undesirable appearance. In particular, it has been found that labels made from polypropylene films having a voided or cavitated layer resulting from the presence of a voiding agent such as chalk acquire a non-uniform or leathery appearance when they shrink. This is particularly disadvantageous as bonding of the molded polymer to the labels is usually improved significantly if the surface of the label which contacts the polymer being molded is voided or cavitated. Furthermore, voided or cavitated films are often preferred for forming labels as they are usually opaque and provide good contrast for printed indicia on the labels.

EP 0312226-A describes oriented films having a core layer of non-voided propylene homopolymer with a layer of voided propylene homopolymer on one surface of the core layer and a printable layer on the other surface of the core layer. Such films are suitable for packaging.

According to the present invention there is provided an in-mold label made from a biaxially oriented polymeric film comprising a core layer of pigmented propylene homopolymer which is non-voided, or is only voided to a degree sufficiently small that a leathery appearance does not result when in-mold labelling is effected, with a layer of voided propylene homopolymer on one surface of the core layer and forming an outer surface of the film and a printable layer on the other surface of the core layer, said pigmented layer containing a pigment having a mean particle size of less than one micron and being selected from titanium dioxide, barium sulfate, zinc sulfide, calcium carbonate, mica and talc, and said voided layer containing sufficient voiding agent that the voided layer bonds to the molded article whilst not producing a leathery effect.

Labels according to present invention have been applied to articles using in-mold labelling techniques without producing the leathery effect referred to hereinbefore.

The present invention further provides articles having labels in accordance with the present invention applied thereto by in-mold labelling.

It is generally preferred that the voided layer should represent a relatively minor proportion of the total thickness of the film so that an unevenness in the voided layer following shrinkage of the film during in-mold labelling has little or no effect on the label as a whole. Typically the voided layer will represent up to 20 percent of the total film thickness, but it can form less than 15 percent or even less than 8 percent of the total film thickness. In general, however, the voided layer should be sufficiently thick that as a result of in-mold labelling, satisfactory adhesion to the rest of the molded article is achieved.

In addition to the thickness of the voided layer, which will usually be from 2 to 10 µm thick, the degree of voiding can affect the properties of the films in inmold labelling. As will be appreciated by those in the art, the degree of voiding of polypropylene by a voiding agent will depend on the particular voiding agent used and the amount of such agent which is used. The partide size of the voiding agent will usually be in the range of from 1 to 15  $\mu m$  and preferably from 3 to 5  $\mu m$ .

The voiding agent can be organic, for example a polyamide (eg a nylon) or a polyester (eg polyethylene terephthalate or polybutylene terephthalate), or inorganic, for example chalk. Chalk is particularly preferred as the voiding agent, the chalk content of the voided layer preferably being up to 5 percent by weight of the layer. The amount of voiding agent in the voided layer will, however, usually be at least 2.5 percent by weight of the layer and it can be up to 15 percent by weight of the layer. Preferred amounts of voiding agent are in the range of from 5 to 10 percent by weight of the layer. Insufficient voiding agent can lead to poor bonding of tabels to the molded articles following in-mold labelling, whereas excessive amounts of voiding agent can lead to the labels acquiring a leathery effect following in-mold labelling.

The voided layer can also contain one or more adhesion promoters which serve to increase adhesion between the voided layer and the polymer to which the label is to be bonded in the in-mold labelling process. Examples of adhesion promoters include polymers containing units derived from propylene and at least one of ethylene and but-1-ene, and blends thereof. As will be appreciated, excessive amounts of adhesion promoter can lead to a reduction in voiding.

The core layer of pigmented polypropylene should not be voided or, if it is voided, it should only be voided to a sufficiently small degree that a leathery appearance does not result when in-mold labelling is effected using such films. The pigment in the core layer should have a mean particle size of less than 1 µm for example about 0.2 µm or less.

The amount of pigment in the core layer will usually be up to 20 percent by weight of the layer, and preferably from 7.5 to 15 percent by weight of the layer.

The printable layer is preferably a layer of a copolymer containing units derived from at least two of propylene, ethylene and but-1-ene, although other printable layers can be used. It is particularly preferred to use a 5 printable layer which can be formed by co-extrusion of the layer with the core layer and the layer which becomes voided following biaxial orientation. A particularly preferred printable layer consists of a copolymer of propylene containing a minor amount of units derived from ethylene.

The printable layer can contain one or more pigments, for example as hereinbefore described for the core layer. The printable layer can alternatively be provided by a suitable coating composition, for example by a coating of an acrylic polymer.

If desired, the printable layer can be subjected to a treatment which increases the surface energy of the printable layer, for example using flame or corona discharge treatment.

Labels used in accordance with the present invention can be of various thicknesses, typically from 30 to 300 µm, for example from 50 to 70 µm. The core layer will usually have a thickness of from 45 to 65 µm, it generally being preferred that the voided layer has a thickness of about 5 µm. The printable layer will typically be about 1.5 um thick.

As will be appreciated, when labels in accordance with the present invention are used to effect in-mold labelling, the voided layer should contact the molding 30 polymer with the printable layer being exposed following the molding operation. The in-mold labelling can be effected by known methods, for example by injection molding of polymer into a mold containing the label, for example in the form of a sheet, into a mold.

Films used to make labels in accordance with the present invention can be prepared using known methods. For example, the three layers can be co-extruded to form a web which is then biaxially oriented, either simultaneously using the bubble process or preferably sequentially, e.g. using heated rollers to stretch the web longitudinally and then a stenter oven to stretch the web in the transverse direction. Although the printable layer is preferably formed by co-extrusion, it can be formed by other methods, for example it can be formed by applying a suitable coating to the core layer either after biaxial stretching or between stretching in the longitudinal and transverse directions.

The following Example is given by way of illustration only.

#### Example

A flat three-layer polymer web was produced by coextruding a core layer of propylene homopolymer containing 10 percent by weight of titanium dioxide of average particle size of about 0.2 µm with a layer of propylene homopolymer containing 5 percent by weight of chalk having an average particle size of 3.5 µm on

one side and a layer of a propylene/ethylene (4 percent by weight of units derived from ethylene) on the other.

The three-layer web was then stretched 4.5 times in the direction of extrusion using heater rollers at 120°C, and thereafter it was stretched 10 times in the transverse direction in a stenter oven at 156°C. The copolymer layer was then subjected to corona discharge treatment and the film was wound up.

The resulting film was 55 µm thick, the core layer being 48.5 µm thick, the chalk-containing layer being 5 μm thick, and the copolymer layer being 1.5 μm thick.

Label-sized pieces of film were then cut from the roll of film and used for in-mold labelling to form margarine tubs, the molding being effected by thermoforming polypropylene sheet 900 µm thick at a temperature of 150°C into a suitable die. The molded pots were then removed from the die.

The labels formed from the film of the present invention showed uniform whiteness without a leathery appearance.

#### Claims

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- An in-mold label made from a biaxially oriented polymeric film comprising a core layer of pigmented propylene homopolymer which is non-voided, or is only voided to a degree sufficiently small that a leathery appearance does not result when in-mold labelling is effected, with a layer of voided propylene homopolymer on one surface of the core layer and forming an outer surface of the film and a printable layer on the other surface of the core layer, said pigmented layer containing a pigment having a mean particle size of less than one µm and being selected from titanium dioxide, barium sulfate, zinc sulfide, calcium carbonate, mica and talc, and said voided layer containing sufficient voiding agent that the voided layer bonds to the molded article whilst not producing a leathery effect.
- A label according to claim 1, wherein the voided propylene homopolymer contains an adhesion promoter.
- A label according to claim 2, wherein the adhesion 45 promoter comprises a polymer containing units derived from propylene and at least one of ethylene and but-1-ene, and mixtures thereof.
- A label according to any one of the preceding 50 4. claims, wherein the voided layer of propylene homopolymer is thinner than the non-voided layer of propylene homopolymer.
  - A label according to any one of the preceding claims, wherein the printable layer comprises a polymer containing units derived from at least two of propylene, ethylene and but-1-ene.

- A label according to any one of claims 1 to 4, wherein the printable layer comprises an acrylic polymer.
- A label according to any one of the preceding claims, wherein the printable layer has been subjected to a treatment to increase its surface energy.
- A labelled article including a label according to any one of the preceding claims which has been applied by an in-mold labelling technique.

### Patentansprüche

- 1. In eine Form einzusetzendes Etikett aus biaxial ausgerichteter Polymerfolie mit einer Kernschicht aus pigmentiertem Propylen-Homopolymer, das nicht oder nur bis zu einem solchen ausreichend kleinen Grad mit Hohlräumen versehen ist, daß kein lederartiges Erscheinungsbild entsteht, wenn man das Etikettieren in der Form durchführt, einer auf einer Fläche der Kernschicht befindlichen Schicht aus Hohlräume enthaltendem Propylen-Homopolymer, die eine Außenfläche der Folie bildet, und einer bedruckbaren Schicht auf der anderen Fläche der Kernschicht, wobei die pigmentierte Schicht ein Pigment enthält, das eine Partikelgröße von weniger als einem um besitzt und aus den Materialien Titandioxyd, Bariumsulfat, Zinksulfid. Kalziumkarbonat. Glimmer und/oder Talkum ausgewählt ist, und die mit Hohlräumen versehene Schicht genügend hohlraumerzeugende Mittel enthält, daß die sie sich mit dem geformten Gegenstand verbindet, ohne dabei einen lederartigen Effekt zu erzeugen.
- Etikett nach Anspruch 1, dadurch gekennzeichnet, daß das mit Hohlräumen versehene Propylen-Homopolymer ein haftungsförderndes Mittel enthalt
- Etikett nach Anspruch 2, dadurch gekennzeichnet, daß das Haftfördermittel ein Polymer enthält, das von Propylen und wenigstens einem der Stoffe Äthylen und But-1-en und Gemischen daraus abgeleitete Einheiten besitzt.
- Etikett nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß die mit Hohlräumen versehene Schicht aus Propylen-Homopolymer dünner als die hohlraumfreie Schicht aus Propylen-Homopolymer ist.
- Etikett nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß die bedruckbare Schicht ein Polymer enthält, das von wenigstens zwei der Verbindungen Propylen, Äthylen und But-1-en abgeleitete Einheiten besitzt.

- Etikett nach einem der Ansprüche 1 bis 4, dadurch gekennzeichnet, daß die bedruckbare Schicht ein Acrylpolymer enthält.
- Etikett nach einem der vorhergehenden Ansprüche, dadurch gekennzeichnet, daß die bedruckbare Schicht einer Behandlung zur Steigerung ihrer Oberflächenenergie unterzogen worden ist.
- Etikettierter Gegenstand mit einem Etikett nach einem der vorhergehenden Ansprüche, das mittels eines in der Form etikettierenden Verfahrens angebracht worden ist.

### 5 Revendications

- 1. Etiquette appliquée dans le moule, réalisée à partir d'un film polymère orienté biaxialement, comprenant une couche formant âme faite d'un homopolymère de propylène pigmenté qui est dépourvu de vides ou qui ne comporte qu'une quantité de vides suffisamment petite pour qu'un aspect du cuir n'apparaisse pas quand un étiquetage dans le moule est effectué, avec une couche faite d'un homopolymère de propylène comportant des vides sur l'une des surfaces de la couche formant âme et constituant une surface extérieure du film, et une couche imprimable sur l'autre surface de la couche formant âme, ladite couche pigmentée contenant un pigment ayant une taille de particule moyenne inférieure à un µm et choisi parmi le dioxyde de titane, le sulfate de baryum, le sulfure de zinc, le carbonate de calcium, le mica et le talc, et ladite couche comportant des vides contenant une quantité suffisante d'agent de création de vides pour que la couche comportant des vides adhère à l'objet moulé tout en ne faisant pas apparaître l'aspect du
- Etiquette selon la revendication 1, dans laquelle l'homopolymère de propylène comportant des vides contient un agent favorisant l'adhérence.
  - Etiquette selon la revendication 2, dans laquelle l'agent favorisant l'adhérence comprend un polymère contenant des unités dérivées du propylène et au moins un élément parmi l'éthylène et le but-1ène, et leurs mélanges.
- 50 4. Etiquette selon l'une quelconque des revendications précédentes, dans laquelle la couche comportant des vides en homopolymère de propylène est plus mince que la couche dépourvue de vides en homopolymère de propylène.
  - Etiquette selon l'une quelconque des revendications précédentes, dans laquelle la couche imprimable comprend un polymère contenant des unités dérivées d'au moins deux éléments parmi le propy-

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lène, l'éthylène et le but-1-ène.

- Etiquette selon l'une quelconque des revendications 1 à 4, dans laquelle la couche imprimable comprend un polymère acrylique.
- 7. Etiquette selon l'une quelconque des revendications précédentes, dans laquelle la couche imprimable a été soumise à un traitement en vue d'augmenter son énergie superficielle.
- Objet étiqueté comprenant une étiquette selon l'une quelconque des revendications précédentes, qui a été appliquée par une technique d'étiquetage dans le moule.

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